

Optical properties of atmospheric aerosols from moments of the particle size distribution

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Abstract. The method of moments is a powerful tool for directly calculating the lower-order moments of an aerosol size distribution without full knowledge of the distribution itself. These moments can be tracked in space and time to represent the effects of nucleation, evaporation and growth, coagulation, and complex flowfield dynamics. This letter applies quadrature methods for efficient estimation of aerosol optical properties directly from a known moment sequence. The approach is demonstrated using particle size distributions obtained from fits to field measurements. Comparison of light-scattering phase functions and other integral optical properties calculated from the moments with those obtained from the full distributions shows that the lower-order moments lead to an efficient and accurate parametrization for these properties.

Introduction

Virtually all properties of atmospheric aerosols and clouds depend strongly on particle size distribution. These include light scattering and absorption, cloud droplet activation, and wet and dry deposition. These properties underlie the major role of aerosols in radiative forcing of climate (both warming and cooling) by directly and indirectly (through clouds) influencing the radiation balance of the atmosphere [Charlson *et al.*, 1992; Lacis and Mishchenko, 1995].

Particle size distributions are shaped by complex nonlinear processes that are difficult to represent in models. These include nucleation of new particles; evaporation, growth and aggregation of existing particles; and transport. A full description would require tracking the evaporation/growth kinetics for each of the billions of discrete particle sizes in the growth sequence, ranging from single condensable vapor molecules, through nucleated clusters, to particles in the optical size range. Fortunately, knowing the full size distribution in such detail is overkill. In standard bin models, particles are generally grouped by number of molecules (g) in geometric proportion (e.g., $g = 2-4, 4-8, 8-16$, etc.). Here a primary difficulty is partitioning the bins to achieve sufficient resolution while keeping the total number of bins manageable for tracking in complex mixing flows. Preservation of adequate resolution to describe the light-scattering effects related to the role of aerosol forcing in climate change, is particularly difficult to achieve in bin-type models. For example, a factor of two uncertainty in g translates to a

factor of four uncertainty in light-scattering cross section in the Rayleigh size regime (particle radius small compared to the wavelength of light).

The method of moments (MOM) permits simulation of aerosol dynamics by tracking the lower-order moments of an aerosol size distribution in space and time without detailed knowledge of the distribution itself [Hulburt and Katz, 1964; McGraw and Saunders, 1984; Pratsinis, 1988]. The k th radial moment is defined as

$$\mu_k = \int r^k f(r) dr \quad (1)$$

where f is the number of particles per unit volume in the size range r to $r + dr$ and the integral is over the full range of particle radius r . Apart from readily determined proportionality constants, μ_0 = particle number density (cm^{-3}), μ_1 = particle radius density (cm/cm^3), μ_2 = particle surface area density (cm^2/cm^3), μ_3 = particle volume fraction (cm^3/cm^3), etc. The present discussion is limited to spherical particles. Extension of the MOM to non-spherical particles characterized by two size coordinates (r_1, r_2) is described by Hulburt and Katz [1964]. In general, the distribution function and its moments are functions of position and time, but to simplify notation we suppress this dependence.

In this letter we introduce a quadrature method for direct calculation of the optical properties of an atmospheric aerosol from a sequence of lower-order moments of its size distribution. Unlike other parameterization approaches, the quadrature method is not based on fits to Mie scattering calculations or on assumed functional forms for the shape of the size distribution. Variable numbers of moments can be used, depending on availability and the level of accuracy required. Finally, although much of the motivation for the present study derives from the availability of moments from aerosol dynamics simulations based on the MOM, the source of the moments is not important to the quadrature method. Thus, if sufficient information is available to infer the moments by other means, such as retrieval from multiwavelength particulate extinction measurements [Livingston and Russell, 1989], the methods developed here can also be readily applied.

Optical Properties

Here we briefly review several important aerosol optical properties while establishing notation for the calculations that follow. The extinction coefficient $k(\lambda)$ gives the loss of light intensity of a primary beam passing through a medium:

$$I = I_0 \exp[-k(\lambda)z] \quad (2)$$

where z is distance measured along the propagation path and λ is wavelength. For single particles it is convenient to express the extinction in terms of the total cross section, $C_{\text{ext}}(r, \lambda)$ or its

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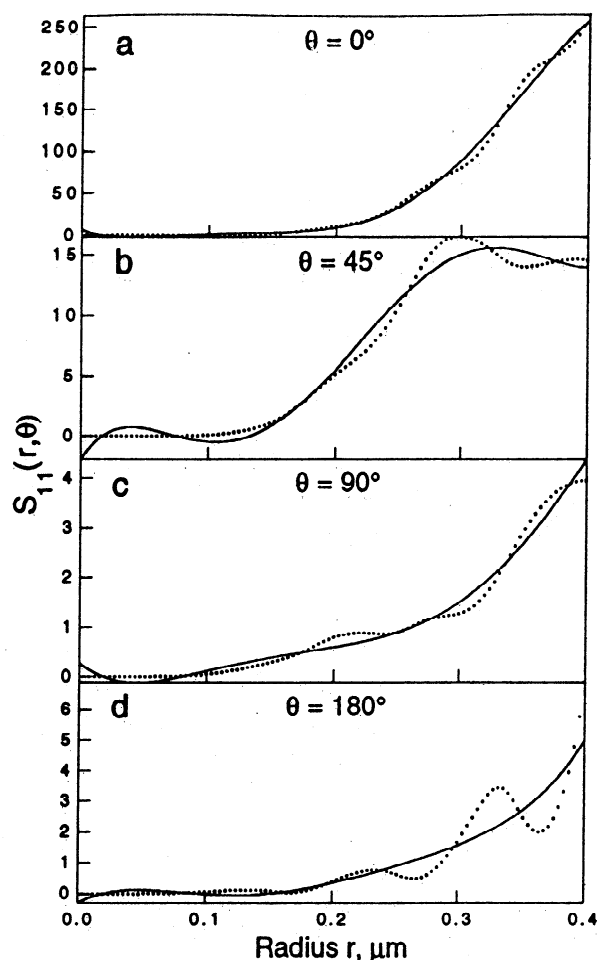


Figure 1. Mie scattering angular distributions (unnormalized). Dotted curves result from Mie scattering calculations at a wavelength of $0.6328 \mu\text{m}$. Solid curves are fifth-degree polynomial fits.

normalized value $Q_{ext}(r, \lambda) = C_{ext}(r, \lambda) / \pi^2$. The extinction coefficient for a distribution of particles, which equals the total cross section per unit volume, is:

$$k(\lambda) = \int C_{ext}(r, \lambda) f(r) dr = \pi \int r^2 Q_{ext}(r, \lambda) f(r) dr \quad (3)$$

The phase function specifies the angular distribution of scattered light and can be written as [Bohren and Huffman, 1983]:

$$p_\lambda(\theta) = N \int S_{11}(r, \lambda, \theta) f(r) dr \quad (4)$$

where $S_{11}(r, \lambda, \theta)$ gives the angular distribution of scattered light from a single particle for unpolarized incident light, the angle θ is measured from the forward scattering direction, and N is a normalization constant.

Equations 3 and 4 each involve integration of a known scattering function weighted by the aerosol distribution. The dotted curves in Fig. 1 show the functions $S_{11}(r, \lambda, \theta)$ evaluated for several angles as a function of particle radius. These were obtained using the Mie scattering code from Appendix A of Bohren and Huffman [1983]. Other optical properties such as the asymmetry factor (\bar{g}_{asym}) and the hemispheric backscatter fraction (β) can be expressed either as integrals over the phase

function [Wiscombe and Grams, 1976] or, upon interchange of the order of integration, equivalently in the general form similar to Eqs. 3 and 4:

$$I = \int \sigma(r) f(r) dr \quad (5)$$

where $\sigma(r)$ is a known optical function. Thus,

$$\bar{g}_{asym} = K^{-1} \int r^2 Q_{sca}(r, \lambda) \bar{g}_{asym}(r) f(r) dr \quad (6a)$$

$$\beta = K^{-1} \int r^2 Q_{sca}(r, \lambda) \beta(r) f(r) dr. \quad (6b)$$

where $Q_{sca}(r, \lambda)$ is the normalized efficiency for scattering. The normalization constant $K = \int r^2 Q_{sca}(r, \lambda) f(r) dr$ is proportional to the total scattering for the distribution and also subject to estimation by the methods to be described.

There is ample precedent for relating the optical properties of an aerosol to the moments of its size distribution. For particles in the Rayleigh size regime $Q_{sca}(r, \lambda) \propto r^4$ and the total scattering cross section is thus proportional to the sixth moment of the size distribution. For particles much larger than the wavelength, $Q_{ext}(r, \lambda) \approx 2$, and the extinction coefficient is thus proportional to the second moment of the size distribution [Bohren and Huffman, 1983]. Chylek (1978) identified an important linear relationship ($Q_{ext}(r, \lambda) \approx cr$), valid under certain wavelength to radius conditions, between the extinction coefficient and the third moment, corresponding to the liquid water content of a cloud or fog; this has been experimentally verified [Gertler and Steele, 1980]. Accurate parameterizations for several radiative properties of water clouds, including the volume extinction coefficient, single-scattering albedo, and asymmetry factor, have recently been developed in terms of the effective particle radius $r_{eff} = \mu_3 / \mu_2$ [Hu and Stamnes, 1993]. Together, these results suggest that at least several of the most important optical properties of an aerosol can be obtained efficiently as byproducts of simulations based on the MOM, which furnish the moments. In the following section we present a new and systematic extension of this approach. In addition to the aforementioned properties, the extended approach can also be used to approximate the angular distribution of scattered light (Eq. 4).

Moments and Quadrature

Gaussian quadrature provides a systematic method for approximate evaluation of integrals of the form given by Eq. 5 while underscoring the importance of the moments [Lanczos, 1988]. For n -point quadrature the result for arbitrary $\sigma(r)$ is:

$$\int \sigma(r) f(r) dr \approx \sum_{i=1}^n \sigma(r_i) w_i \quad (7)$$

where the abscissas r_i and weights w_i satisfy the $2n$ moment conditions:

$$\mu_k = \int r^k f(r) dr = \sum_{i=1}^n r_i^k w_i \quad (8)$$

for $k = 0$ through $2n - 1$. Although in principle one can solve for the abscissas and n weights from the set of $2n$ equations (Eqs.

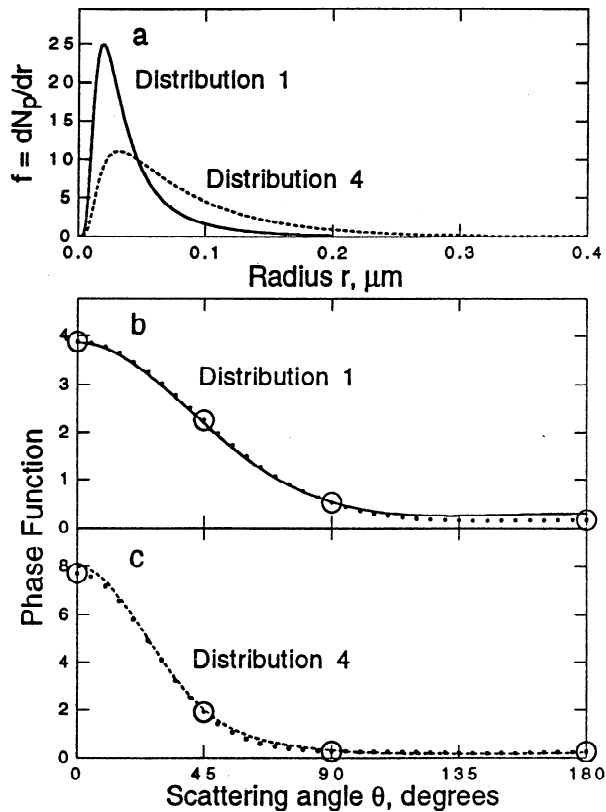


Figure 2. a. Normalized test aerosol size distributions labeled as in the original paper [Hoppel et al., 1990]. Solid curve, distribution 1; dashed curve, distribution 4. b. Phase function for distribution 1. Smooth curve, from numerical integration of Eq. 4 using the full size distribution; points, quadrature approximation, computed at 5° increments, using six moments. Open circles mark angles corresponding to the angular distributions shown in Figure 1. c. Similar results for distribution 4. Normalization for the phase functions has been chosen such that the integral over all scattering directions is 4π .

8), much more efficient approaches are available, which will be described in a future paper. For the special case that $f(r) = \exp(-r)$ the abscissas and weight factors are the same as for Laguerre integration, but more generally $f(r)$ is of nonclassical form [Press and Teukolsky, 1990]

For the present study, the most important feature of the quadrature approximation is that the abscissas and weights do not depend on the full distribution function, $f(r)$, but only on its lower order moments. Thus we can obtain approximations to the integral of Eq. 5 without requiring the full distribution function itself. An additional, very convenient property is that the abscissas and weights are independent of the optical kernel function $\sigma(r)$.

For the special case that $\sigma(r)$ is a polynomial function of r of order $\leq 2n-1$:

$$\sigma_{2n-1}(r) = a_{2n-1}r^{2n-1} + a_{2n-2}r^{2n-2} + \dots + a_1r + a_0. \quad (9)$$

where a_i are arbitrary coefficients, it is well known that Eq. 7 is exact [Lanczos, 1988]. Thus polynomial fits to $\sigma(r)$ can be introduced as a straightforward generalization of powers-of- r type approximations, noted above for the total scattering cross

section. The solid curves in Fig. 1 show least-squares, fifth-degree polynomial fits to the corresponding Mie points over the radius range $0-0.4 \mu m$. The main difficulty in doing such a fit lies in selecting the optimal fit range, which will depend on the size range of the aerosol distribution itself. Nevertheless, the fact that quadrature is exact for polynomial functions provides a guideline for selecting the order of quadrature and for predicting the accuracy of the method. Alternatively, evaluation of the right-hand side of Eq. 7, which is the method used in the sample calculations below, is more direct in that polynomial fits to $\sigma(r)$ are not required.

Results and Discussion

The methods of the previous section are demonstrated using test particle size distributions obtained from field measurements. Figure 2a shows two normalized marine aerosol size distributions [Hoppel et al., 1990], which following these investigators has been smoothed by fitting a curve of the form

$$\ln[f(r)] = C_0 + C_1(\ln r) + C_2(\ln r)^2 + \dots \quad (10)$$

to the measured values of dN_p/dr . We set the refractive index of the particles at 1.55, independent of particle size, and $\lambda = 0.6328 \mu m$. The phase functions can be computed directly from Eq. 4, as the size distributions are known, and are shown as the solid curves in Figs. 2b and 2c. Similarly, the moments are computed via numerical integration over the smoothed distributions.

The method illustrated here for 3-point and 4-point quadrature can be extended to higher order if required. For the phase function we use only 3-point quadrature. Accordingly, we require here only the six lowest-order moments $\{\mu_0, \dots, \mu_5\}$ for each size distribution. The abscissas $\{r_i\}$ and weights $\{w_i\}$, for $i = 1$ thru 3, are obtained from the moments (see discussion following Eq. 8), and the quadrature approximation to the phase function follows immediately from Eqs. 4 and 7,

$$p_\lambda(\theta) \equiv N \sum_{i=1}^3 S_{11}(r_i, \lambda, \theta) w_i. \quad (11)$$

Evaluating the right-hand side of Eq. 11 for the respective test distributions gives the points shown in Figs. 2b and 2c. The results are in quite good agreement with the solid curves obtained numerically from the full test distributions. Results for additional optical properties, including the 180° backscattering cross section:

$$\sigma_b = \pi \int r^2 Q_b(r) f(r) dr, \quad (12)$$

where $Q_b(r)$ is the efficiency for back scattering from a particle of radius r [Bohren and Huffman, 1983], are compared in Table 1. Exact results for $k(\lambda)$ and σ_b were obtained numerically from the Mie code of Bohren and Huffman [1983], and for \bar{g}_{asym} and $\bar{\beta}$ from integrals over the phase function given by Wiscombe and Grams [1976] and, with equivalent results, from Eqs. 6. As shown above, each of these quantities is readily expressed in the standard form of Eq. 5 for quadrature approximation; results for 3-point (six moment) and 4-point (eight moment) quadrature are shown in the Table. The large-angle backscatter calculations serve to illustrate the limits of the

Table 1. 3-point (4-point)-quadrature results for optical properties of the test distributions over the size ranges shown in Fig. 2a: $k(\lambda)$, extinction coefficient; g_{asym} , asymmetry parameter; β , hemispheric backscatter fraction; σ_b , 180-degree backscattering cross section.

| | Distribution 1 | | | Distribution 4 | | |
|-------------------------------|----------------|----------|---------|----------------|----------|---------|
| | exact | quad | % error | exact | quad | % error |
| $k(\lambda)$ | 19.353 | 18.935 | -2.2 | 11.040 | 10.498 | -4.9 |
| $\mu\text{m}^2 / \text{cm}^3$ | | (19.829) | (+2.5) | | (11.502) | (+4.2) |
| $\overline{g_{\text{asym}}}$ | 0.4809 | 0.5147 | +7.0 | 0.6379 | 0.5949 | -6.7 |
| | | (0.4931) | (+2.5) | | (0.6485) | (+1.7) |
| β | 0.3172 | 0.2939 | -7.3 | 0.2487 | 0.2397 | -3.6 |
| | | (0.3248) | (+2.4) | | (0.2657) | (+6.8) |
| σ_b | 5.789 | 3.232 | -44.2 | 2.249 | 2.645 | +17.6 |
| $\mu\text{m}^2 / \text{cm}^3$ | | (6.189) | (+6.9) | | (2.915) | (+29.6) |

method. Note that as functions of r , $\pi^2 Q_b(r) = 4\pi S_{11}(\theta = 180^\circ, r)/|k|^2$ where $|k|$ is the magnitude of the wavevector of light in the medium (air). Therefore, the limitations of the quadrature method, seen for σ_b in Table 1 and in Figs. 2b and 2c at 180° , are not independent. In this case the scattering function exhibits large amplitude oscillations (Fig. 1d) that are difficult to sample with just a few abscissas and weights. Nevertheless, even in this extreme case the agreement is significantly improved, in many practical applications, by averaging over wavelength (e.g., the solar spectrum or the bandpass of a satellite borne radiometer).

The quadrature approaches developed here, both direct and through polynomial approximation to $\sigma(r)$, form valuable additions to the limited repertoire of methods available for estimation of aerosol properties from moments of the particle size distribution. The results presented here illustrate the accuracy and power of this technique suitable for evaluating aerosol optical and radiative properties in a wide variety of applications.

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